

Feshbach Resonance and Hybrid Atomic/Molecular BEC-Systems

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Abstract

The interactions responsible for Feshbach resonances in binary atom collisions produce a condensate of molecules in the atomic BEC-system. We discuss the ‘ground state’ of the condensate system and illustrate that its properties differ quantitatively and qualitatively from the properties of a single condensate with atom-atom interactions described by the effective scattering length that characterizes the binary atomic collisions. We show, for example, that the tunneling of atom pairs between the atomic and molecular condensates lowers the energy and can bind the condensate system, giving it the liquid-like property of a self-determined density.

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The recently observed atomic Bose-Einstein condensates [1] are dilute superfluid quantum gas systems and their many-body properties are highly interesting. In contrast to the situation in the traditional (Helium) superfluids, the strength of the inter-particle interactions in the atomic condensates can vary over a wide range of values. In particular, the scattering length that characterizes the atomic interactions can be negative, corresponding to an effective inter-atomic attraction. A condensate of such bosons collapses in the absence of an external potential, but a trapping potential may hold a small negative scattering length condensate in a metastable state [2] that can tunnel [3]. Moreover, the remark that the scattering length may be varied continuously for binary collisions seemed to promise a ‘tunable’ interaction strength. The proposed schemes [4] to change the scattering length involve a resonant scattering process with an intermediate bound molecular state of the interacting atoms. In the scheme that involves a Feshbach resonance [5], recently achieved experimentally in an atomic BEC-system [6], the molecular intermediate state energy is tuned near-resonance (i.e. near the continuum of the incident atom channel) by varying an external magnetic field.

In this letter, we point out that the interactions responsible for the Feshbach resonance in the binary atom collisions produce an additional condensate of molecules. Near-resonance, the properties of the hybrid atomic/molecular condensate differs qualitatively from the properties of a single condensate. As a particularly striking illustration, we will show that the tunneling of atomic pairs between the atomic and molecular condensates can produce a bound many-body state with the liquid-like property of a self-determined density.

The binary atom Feshbach resonances studied by varying a strong external magnetic field in an alkali-atom trap are hyperfine-induced spin-flip processes that bring the colliding atoms to a bound molecular state of different electron spin. The interaction of two alkali atoms, their spins aligned by the magnetic field, is, to lowest order, described by the triplet molecular potential. If the magnetic field is tuned near-resonance, the hyperfine interaction of the electronic (**s**) and nuclear (**i**) spins, $V_{hf} \sim \mathbf{s} \cdot \mathbf{i}$, can simultaneously flip the electronic and nuclear spins of one of the colliding atoms, bringing the molecular system to a bound

vibrational eigenstate of a singlet potential of nearly the same energy as the continuum of the incident atom channel. For the ultra-cold condensate systems, the matrix element for the transition of the initial state of atoms 1 and 2 in spin state $|S_{\text{in}}\rangle$ to the bound molecular state of spin $|S_{\text{m}}\rangle$, can be taken to be independent of the momenta of the atoms. This matrix element, α , is equal to $\alpha = \langle S_{\text{in}} | V_{hf}(1) + V_{hf}(2) | S_{\text{m}} \rangle \times \int d^3x \psi_m^*(\mathbf{x})\psi(\mathbf{x})$, where \mathbf{x} denotes the relative position of atoms 1 and 2 and ψ_m and ψ represent the bound state and continuum wave functions of the initial and intermediate molecular states. The continuum wave function is normalized by requiring ψ to go over into the partial s-wave component of a plane wave at large separation. The hyperfine-induced spin-flips are then described by the following Feshbach resonance contribution to the Hamiltonian:

$$H_{FR} = \alpha \int d^3r \hat{\psi}_m^\dagger(\mathbf{r})\hat{\psi}_a(\mathbf{r})\hat{\psi}_a(\mathbf{r}) + h.c., \quad (1)$$

where $(\hat{\psi})$ and $(\hat{\psi}^\dagger)$ represent the usual annihilation and creation field operators of the atoms $(\hat{\psi}_a, \hat{\psi}_a^\dagger)$ and the molecules in the bound vibrational state $(\hat{\psi}_m, \hat{\psi}_m^\dagger)$.

For the binary atom system, H_{FR} gives a resonant contribution to the atom-atom interaction strength λ_a (where λ_a is proportional to the scattering length a_s , $\lambda_a = 4\pi\hbar^2 a_s/m$): $\lambda_{\text{eff}} = \lambda_a - 2\alpha^2/\epsilon$, where ϵ is the energy of the intermediate molecular state relative to the continuum of the incident atoms. The value of ϵ , which we shall refer to as the ‘detuning’, linearly depends on the magnetic field H . To see that, we note that the energy difference Δ of the singlet and triplet continuum levels, which is the energy gain experienced by a single atom in the spin flip process, is proportional to H : $\Delta = \mu' H$. The detuning is the difference of the Δ and the binding energy E_b of the resonant singlet molecule, so that $\epsilon = \mu'(H - H_0)$, where $H_0 = E_b/\mu'$ is the resonant magnetic field. The expression for λ_{eff} is most easily obtained by determining the energy shift of two atoms with the same momentum, confined to a volume Ω . In first-order perturbation, the atom-atom interaction term, $[\lambda_a/2]\hat{\psi}_a^\dagger\hat{\psi}_a^\dagger\hat{\psi}_a\hat{\psi}_a$, contributes λ_a/Ω to the energy shift ΔE . In second-order perturbation theory, H_{FR} couples the initial two-atom state $|\text{ini}\rangle$ of energy E_{ini} to the intermediate molecular state $|\text{int}\rangle$ of energy E_{int} , contributing $|\langle \text{ini} | H_{FR} | \text{int} \rangle|^2/[E_{\text{ini}} - E_{\text{int}}] = [-2\alpha^2/\epsilon]/\Omega$. The

overall shift is $\Delta E = \lambda_{\text{eff}}/\Omega$, where $\lambda_{\text{eff}} = \lambda_a - 2\alpha^2/\epsilon$ [7].

In the binary collision, the intermediate state is virtual since it does not satisfy energy conservation. In the many-body system, two particle energy conservation considerations do not apply and we argue that, under the appropriate conditions, H_{FR} produces a condensate of molecules. If all atoms are in the same center-of-mass state, then the molecules created in the Feshbach resonance also occupy a single center-of-mass state, the characteristic of a condensate [9]. We can describe the dynamics of the atomic and molecular condensate fields, ϕ_a and ϕ_m , with an effective Hamiltonian-density [10]:

$$\begin{aligned} & \phi_a^* \left[-\frac{\hbar^2 \nabla^2}{2m} + \frac{\lambda_a}{2} |\phi_a|^2 \right] \phi_a + \phi_m^* \left[\frac{-\hbar^2 \nabla^2}{4m} + \epsilon + \frac{\lambda_m}{2} |\phi_m|^2 \right] \phi_m \\ & + \lambda |\phi_a|^2 |\phi_m|^2 + \alpha [\phi_a^2 \phi_m^* + \phi_a^* \phi_m^2] , \end{aligned} \quad (2)$$

where the \mathbf{r} -dependence of the ϕ -fields is understood, and where λ_a , λ_m and λ denote the strengths of the atom-atom, molecule-molecule, and atom-molecule interactions. Having produced the molecular condensate, H_{FR} describes tunneling of atom pairs between the atomic and molecular condensates. In the tunneling, the total number of atomic particles is conserved if we count each molecule as two atomic particles. We find it useful to introduce the atomic particle density $n(\mathbf{r}) = n_a(\mathbf{r}) + 2n_m(\mathbf{r})$, where $n_a = |\phi_a|^2$ and $n_m = |\phi_m|^2$ are the atomic and molecular densities. Note that the effective Hamiltonian depends on the phase difference of ϕ_a^2 and ϕ_m . In describing the static situation, choosing ϕ_a to be real and positive, and noting that the H_{FR} -contribution is minimal when ϕ_m is real, it follows from Eq.(2) that we can describe the statics of a homogeneous condensate system with the following energy density:

$$u = \frac{\lambda_a}{2} n_a^2 + \frac{\lambda_m}{2} n_m^2 + \lambda n_m n_a + \epsilon n_m + 2\alpha n_a \phi_m . \quad (3)$$

To minimize the energy, we introduce a variable x , which is a scaled molecular field, $\phi_m = x\sqrt{n/2}$. The square of x then represents the fraction of atoms that have been converted to molecules, $n_a = n(1 - x^2)$ and the energy per particle, $e = u/n$, reduces to

$$e = \frac{\lambda_a}{2} n + \alpha \sqrt{2n} x + \{[-\lambda_a + \lambda/2]n + \epsilon/2\} x^2 - \alpha \sqrt{2n} x^3 + [\lambda_a/2 - \lambda_m/4 - \lambda/2] n x^4 . \quad (4)$$

Minimizing e with respect to the molecular field parameter x within the interval $(-1, +1)$ determines the equilibrium value of the molecular field for a given atomic particle density n . Taking α to be positive [11], we reduce the problem to the minimization of the dimensionless function $f(x) = \left(u - \frac{\lambda_a}{2}n\right)/\alpha\sqrt{2n}$,

$$f(x) = x + \epsilon'x^2 - x^3 + \beta x^4 . \quad (5)$$

Here $\epsilon' = \{\epsilon + n[-2\lambda_a + \lambda]\}/2\alpha\sqrt{2n}$, and $\beta = n[\lambda_a + \lambda_m/4 - \lambda]/2\alpha\sqrt{2n}$.

A limit of particular interest is that of off-resonant detuning: $\epsilon \gg 2\alpha\sqrt{2n}$, and ϵ exceeding the single particle interaction energies, $\epsilon \gg \lambda_a n, \lambda_m n, \lambda n$. Then $f(x) \approx x + \epsilon'x^2$, with a minimum at $x = -(1/2\epsilon')$, giving a molecular density $n_m \approx n^2[\alpha/\epsilon]^2$. The ground state energy per particle is equal to $u \approx n\lambda_{\text{eff}}/2$, where λ_{eff} is the effective interaction strength in the binary collision picture, $\lambda_{\text{eff}} = \lambda_a - 2\alpha^2/\epsilon$. Thus the off-resonant limit ($\epsilon \gg 0$) gives the same result as the effective scattering length description.

In contrast, as ϵ is lowered near and ‘below’ resonance (i.e. $\epsilon \rightarrow 0$ or $\epsilon < 0$), the condensate system does *not* exhibit resonance features. Instead, as the detuning is lowered in a condensate of fixed density [8], the fraction of molecules increases until at $\epsilon = -2\alpha\sqrt{2n} + n[\lambda - \lambda_m/2]$ [12], all atoms are converted to molecules.

The Feshbach-resonance parameter α , proportional to a molecular overlap matrix element, can take a wide range of values. Calculations for the Feshbach resonances reported at MIT, indicate that $\alpha\sqrt{2n}$ was of the same order of magnitude as $\lambda_a n$. To be definite, we will discuss a system of density $n_0 \sim 10^{14} - 10^{15} \text{ cm}^{-3}$ with $\alpha\sqrt{2n_0} = 5\lambda_a n_0$, roughly corresponding to the Feshbach resonance of largest width detected in the MIT experiments [6]. If $\alpha\sqrt{2n}$ significantly exceeds the interaction energies $\lambda_a n, \lambda_m n, \lambda n$, then $|\beta| \ll 1$ and the fourth order term βx^4 of the f -function of Eq.(5) may be neglected (except when the system approaches the purely molecular condensate). Minimizing $f(x) \approx x + \epsilon'x^2 - x^3$, we obtain a simple analytical expression for the molecular field parameter x :

$$x = [\epsilon' - \sqrt{\epsilon'^2 + 3}]/3 . \quad (6)$$

With this result, we obtain analytical expressions for such quantities as the chemical potential, $\mu = (\partial u / \partial n)$, pressure, $P = n^2 \partial e / \partial n$, sound velocity etc... For example, in this approximation, $\beta = n[\lambda_a + \lambda_m/4 - \lambda]/2\alpha\sqrt{2n} \approx 0$, we obtain

$$P = \frac{\lambda_a n^2}{2} + n \frac{\alpha\sqrt{2n}}{9} [\epsilon' - \sqrt{\epsilon'^2 + 3}] \left[1 - \frac{\epsilon'}{3}(\epsilon' - \sqrt{\epsilon'^2 + 3}) \right] + n \frac{(\lambda - 2\lambda_a)n}{18} [3 + 2\epsilon'^2 - 2\epsilon'\sqrt{\epsilon'^2 + 3}]. \quad (7)$$

In Fig. (1) we show this pressure for fixed density $n = n_0$, and $\lambda = 2\lambda_a$ and $\lambda_m = 4\lambda_a$ (implying $\beta = 0$, so that Eq.(6) is exact), as a function of the detuning.

The negative pressure near zero detuning indicates that the condensate system increases its density. We consider a condensate that is confined by fixed ‘walls’ to a well-defined volume that is large enough to describe the condensate by the above theory (which is an infinite system treatment). Lowering the detuning to a value of negative pressure, causes the system to occupy a volume less than that allowed by the walls. In the effective scattering length description, which gives negative pressure when $\epsilon < 2\alpha^2/\lambda_a$, that volume is zero: the system collapses. In contrast, the hybrid atomic/molecular condensate takes on a *finite* volume, determined by the density that minimizes the energy e . In Fig.(2) we plot e as a function of the relative atomic particle density $n_s = n/n_0$ for different values of the scaled detuning $\epsilon_s = \epsilon/\alpha\sqrt{2n_0}$ for the same parameters as Fig.(1). At $\epsilon_s = 5$, for which the effective scattering length description predicts zero pressure, the condensate system still gives a positive pressure and e is a monotonically increasing function of the density. Without the confining walls the condensate expands indefinitely as a gas. At $\epsilon_s = 4.5$, the pressure is still positive at $n = n_0$, but the energy is negative at small densities, implying that the many-body system is bound. The binding stems from the atom pair tunneling contribution $2\alpha n_a \phi_m$ to the energy density. At $\epsilon_s = 4.5$, the energy e reaches a minimum at density less than n_0 . Thus, without walls, the condensate expands until its density reaches the value that minimizes e . Since that density requires a volume greater than that allowed by the walls, a confined condensate keeps its density. At $\epsilon = 3.8$, on the other hand, the pressure is negative at $n = n_0$ and the density that minimizes the energy exceeds n_0 : the condensate spontaneously decreases its

volume to a fraction of the volume of confinement. The condensate finds its own volume and displays the liquid-like property of a self-determined density. Interestingly the value of this density (or, alternatively, the volume) is determined by the detuning and can be varied by changing the magnetic field.

In Fig.(3), we show the trajectory that the condensate follows in the plane of the detuning and density, as the detuning is lowered adiabatically through the Feshbach resonance. The condensate is gaseous and occupies all of the allowed volume – its density remains equal to n_0 until its pressure vanishes. Lowering the detuning further, the condensate contracts and subsequently expands along the curve of zero pressure. Note that for a range of negative detuning values, we find two densities at which the pressure vanishes. The lowest density at which this happens actually corresponds to a maximum in the density, rather than a minimum, and the system could not remain on this part of the curve. Before the $P = 0$ curve ‘turns back’, the density n discontinuously changes to its initial value, corresponding to an abrupt expansion of the condensate to fill the maximum allowed volume. For an atomic-trap with a smooth trapping potential, instead of ‘walls’, we expect that a similar contraction and subsequent expansion.

The extent to which these very interesting phenomena can be observed crucially depends on the relevant time scales. Since the Feshbach resonances create molecules in high vibrational states, the molecular lifetime, τ_m , is significantly shorter than the atomic lifetime. The decay rate, τ_m^{-1} , is proportional to the atomic and molecular densities, $\tau_m^{-1} = c_{ma}n_a + c_{mm}n_m$, where the c -coefficients are the rate coefficients due to molecule-atom (c_{ma}) and molecule-molecule (c_{mm}) collisions. Estimates based on calculations for hydrogen give molecular rate coefficients $\sim 10^9 - 10^{11} \text{ cm}^3/\text{sec}$ [13]. Using the lower estimate, a condensate of pure molecules of density 10^{14} cm^{-3} lives for 10^{-3} sec . With the relevant time scale for the condensate dynamics $\hbar/\lambda n \sim 10^{-5} \text{ seconds}$, it might be possible to adiabatically tune ϵ near resonance, although these estimates clearly underline the need for calculations of these quantities. Off-resonance ($\epsilon > 0$), only a small fraction of atoms is converted to molecules, and, although the molecules decay by colliding with the remaining atoms, the condensate

is constantly being replenished by tunneling from the atomic condensate. In this limit, $n_m \approx n_a^2 |\alpha/\epsilon|^2$ and particle loss may be described by a rate coefficient that is the usual atomic rate coefficient, c_{aa} , enhanced with the rate at which atom pairs which have been converted to molecules are being lost to molecule-atom collisions: $dn/dt \approx -c_{eff}n^2$, with $c_{eff} = c_{aa} + 2c_{am}n|\alpha/\epsilon|^2$.

In conclusion, we have pointed out that the Feshbach resonance interactions can produce a condensate of molecules in an atomic BEC system. Off-resonance, many properties of the condensate system are described correctly in the effective scattering length description. However, this description does not predict the small molecular condensate, which could be kept for times exceeding the lifetime of a pure molecular condensate. Near-resonance, the tunneling between the atomic and molecular condensates can dramatically alter the properties of the condensate systems, a fact that we have illustrated by discussing the appearance of a condensate phase with self-determined density.

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- [6] S. Inouye et al., *Nature*, **392**, 125 (1998).
- [7] A more rigorous derivation (see, e.g., Ref. [5]) leads to the same dispersive ϵ^{-1} -dependence.
- [8] We assume here that $\beta < -1$. Otherwise a discontinuity in x may occur as we shall discuss in a more comprehensive treatment.
- [9] In a more systematic approach, one may take the expectation value of the Heisenberg equation for the field-operators. Denoting the fields by $\phi = \langle \hat{\psi} \rangle$, the molecular field $\phi_m = \langle \hat{\psi}_m \rangle$ is ‘driven’ by a source term $\alpha \langle \hat{\psi}_a \hat{\psi}_a \rangle \approx \alpha \phi_a^2$ and ϕ_m does not vanish.
- [10] This effective Hamiltonian may be derived from a Gaussian Approximation treatment such as discussed in A. Kerman and P. Tommasini, *Ann. Phys. (N.Y.)*, **260**, 250 (1997).
- [11] Negative α is equivalent to replacing x by $-x$

[12] Calculating the energy gain of converting molecules to atoms in a purely molecular condensate, we obtain $\epsilon + 2\alpha\sqrt{2n} - n[\lambda - \lambda_m/2]$ per molecule that is converted. If this energy is positive, the molecular condensate lowers its energy by producing atoms.

[13] A. Dalgarno, private communication.

Figure Captions

Fig.1 : Scaled pressure $P_s = P/[\lambda_a n_0^2/2]$ as a function of the scaled detuning $\epsilon_s = \epsilon/[\alpha\sqrt{2n_0}]$.

The dashed curve shows the effective scattering length prediction.

Fig.2 : Plot of the energy per particle, $e_s = e/[\lambda_a n_0/2]$, as a function of the scaled density $n_s = n/n_0$ for the detunings shown on the figure. Negative e_s implies a bound many-body state.

Fig.3: Plot illustrating the density variation of a condensate, confined by fixed walls, as the detuning $\epsilon_s = \epsilon/[\alpha\sqrt{2n_0}]$, is adiabatically lowered through the Feshbach resonance. For the lowest initial density, the system contracts and expands along the curve of zero pressure, shown in dashed line. Before returning to its initial density, the system undergoes an abrupt change in the density. Similarly, the system of medium initial density contracts and expands but does not change its density abruptly. The density of the system of highest initial density remains constant.

Fig. 1

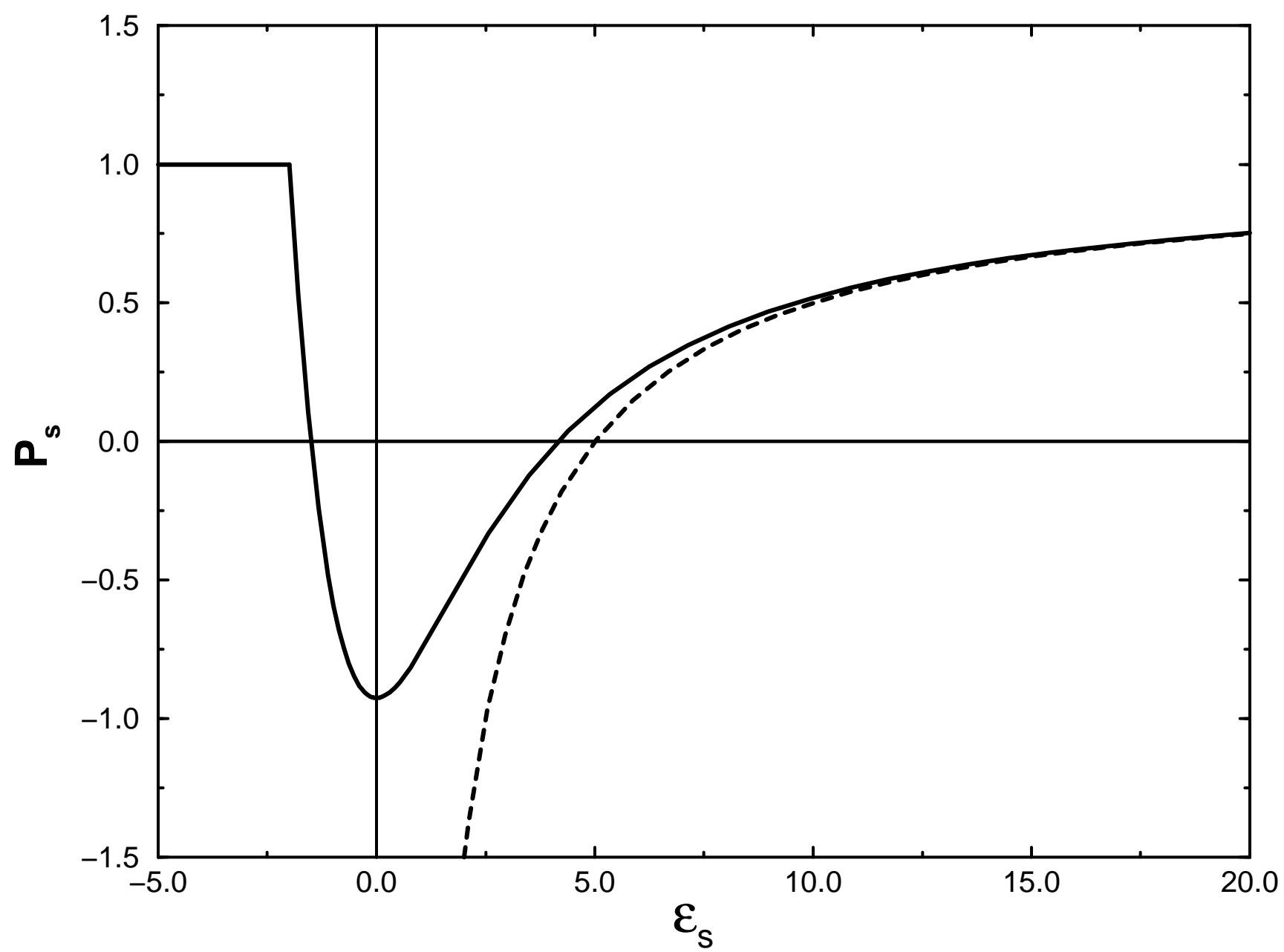


Fig. 2

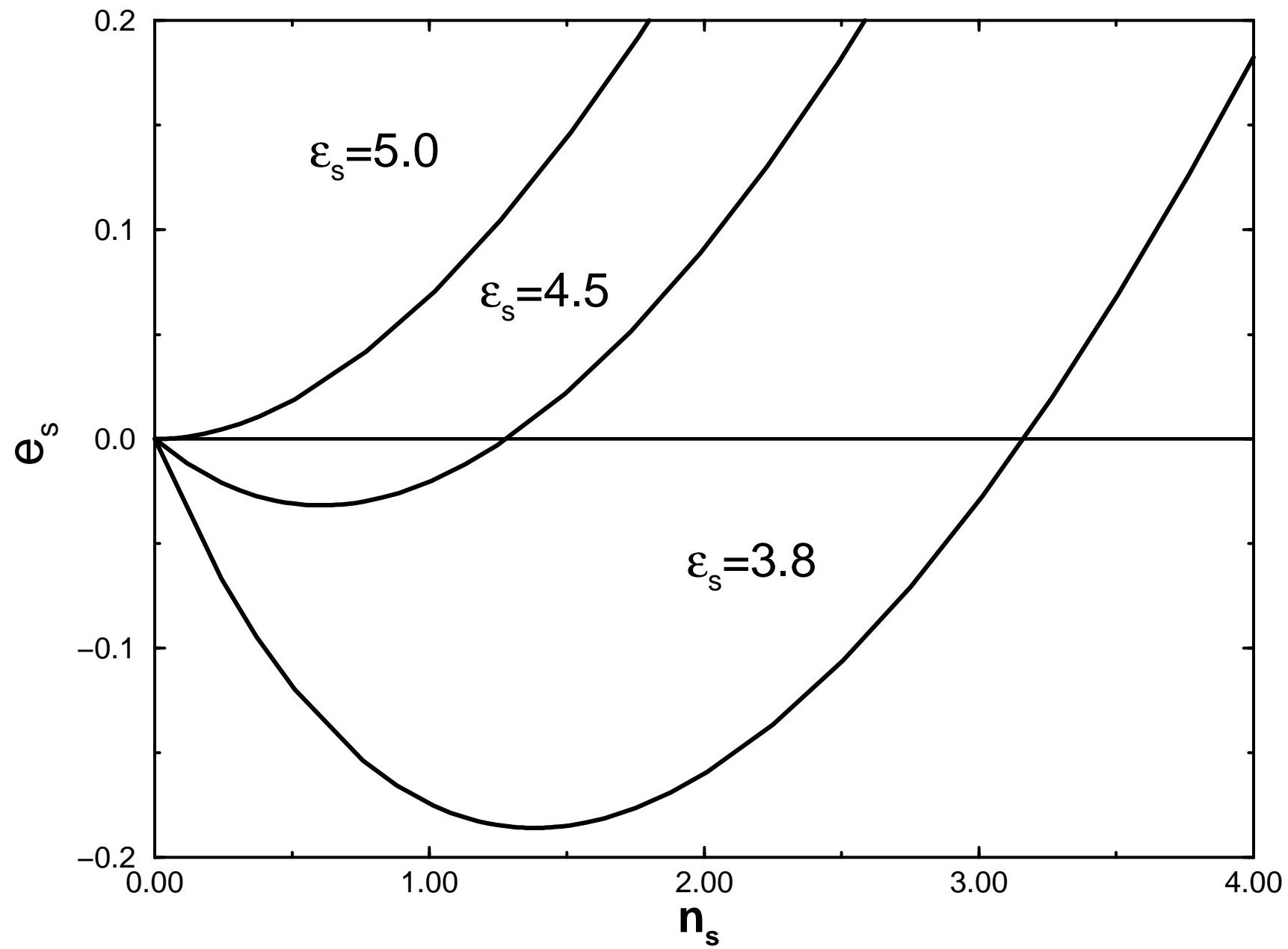


Fig. 3

